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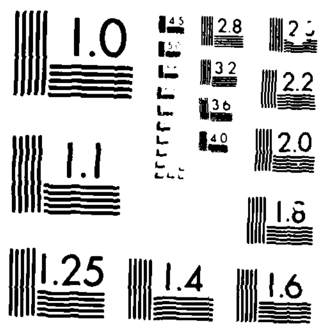

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Interim Technical Report

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Experimental Study of Electronic States at Metal-Dielectric Interfaces

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Because infrared surface plasmons on metal surfaces propagate for many wavelengths, a measurement of the transport of these excitations can be used to monitor the surface itself. Using the metal edge with a broadband continuous source. Measurements of the transmission of IR surface plasmons across metal surfaces covered with a chemisorbed atomic monolayer or with thin dielectric or molecular films have been carried out and, compared with those obtained by reflection spectroscopy.

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I. ABSTRACT

New infrared techniques are being used to explore both the time and frequency domain spectra of electronic defect and interface states in semiconductors.

II. PROJECT OBJECTIVES

The development of new infrared techniques which are ideally matched to the interface problem will continue. These surface plasmon (SP) techniques will be used to measure and characterize the physical properties of electronic states at metal-dielectric interfaces. A closely related objective is to explore the near field electrodynamics of structures which produce evanescent fields. At the present time the efficient SP coupling properties of fine grating structures on metal surfaces are of particular interest. The generation of surface plasmons on single crystal metal surfaces would permit the spectroscopic study of controlled oxide growth and the generation of SP's at metal semiconductor interfaces would provide a starting point for interface photoconductive spectroscopy. Finally it is proposed that high resolution IR laser spectroscopy by carried out on thin films of deep level electronic impurities which are produced in bulk semiconductors by ion implantation. One object of such an investigation would be to measure the efficiency of spectral hole burning for possible application to frequency domain optical storage.

III. ACCOMPLISHMENTS

During the past year the optical and physical properties of propagating infrared surface plasmons on metal substrates have been investigated.

We have found that the key parameter to understanding the optical properties of infrared surface plasmons (SP's) is the height of the wave above the metal surface. SP's are unique among E & M bound modes in that they are not confined to a particular region of space which is defined by physical boundaries, as are the bound modes in a dielectric waveguide or transmission line. SP's are bound to an interface and determine their own degree of confinement based on the intrinsic physical properties of the interface supporting them. The height turns out to be extremely sensitive to very small changes in the SP phase velocity which we can directly measure.

We have found that at the edge of a dielectric coating on a metal substrate, strong linear coupling occurs between the SP and plane E & M waves. We have shown how this coupling can be used to obtain interference between the SP and plane wave modes so that the SP phase velocity can be measured interferometrically. This SP interferometer has been used to measure the infrared electron mass of Au and Ag at 10 μm wavelength.

The wave height also determines the efficiency with which the incident radiation excites the SP's. We have shown experimentally that at about 100 μm wavelength (where the height is larger) SP excitation is inefficient and plane waves dominate the surface transmission signal. At 10 μm wavelength (where the height is small) the results of the SP interferometer measurements are used to conclude that only the SP mode contributes to the surface transmission signal for bare or uniformly coated surfaces. These results allow us to correctly interpret SP attenuation measurements at both wavelengths. For bare metals at 100 μm we show that previous reports of anomalously large SP attenuation are in error. By combining both the experimental and theoretical results an understanding of

SP coupling and propagation is developed.

We have also demonstrated the use of broadband infrared SP transmission to study molecules on metal surfaces. SP's were excited by an incoherent (Nernst glower) source using a dispersion compensating edge coupler developed specificially for this purpose. We found that the SP's are an order of magnitude more sensitive to surface phenomena than plane wave reflection measurements. The SP technique was signal-to-detector-noise limited and in order to be competitive with existing vibrational surface spectroscopy techniques (EELS and SRS) a higher throughput, UHV compatible SP coupling technique is required. Broadband SP's may prove useful to study the broader spectral features associated with the effects of roughness, surface contamination, interface states or variations in the optical conductivity of the metal which supports the SP.

A search for evidence of electron-SP scattering in Ag produced inconclusive results. With the appropriate dielectrics, however, metal-dielectric interface reflectance measurements should be able to determine the existence and magnitude of electron-SP scattering in noble metals.

More successful was our study of the dielectric function of silver by SP's. We observed for the first time changes in the effective complex dielectric function of silver with the refractive index of the dielectric half space. These results were obtained by the excitation of SP's at several wavelengths in the visible with attenuated total reflection experiments. Our results with different glass substrates and liquids with varying refractive indices will be interpreted in terms of modifications to the standard free electron model.

We also found that a multi-mode Lorentz oscillator model can be used to study the effects of the depolarization field on vibrational modes of thin film systems with multiple molecular sites or with several IR active modes. For strong modes, line narrowing and loss of structure occur as coverage increases. Spectral linewidths at high coverages are found to be homogeneous, despite substantial inhomogeneity in the adsorbate system at the molecular level. Absorption line intensity is transferred from lower to higher frequency modes through a mutual screening and antiscreening process. The positive polarizability associated with the higher frequency mode (at the frequency of the lower mode) screens the lower mode, whereas the negative polarizability associated with the lower mode (at the frequency of the higher mode) antiscreens the higher mode. Thus we find that the depolarization field can distort the correspondence between spectral features and single molecule properties.

Finally an interesting new effect has been observed during our measurement of the SP attenuation coefficients for bare and coated Ag and Au metals in the 10 μm wavelength region. For the bare metal agreement is obtained with the Drude model predictions but when these same surfaces are coated with germanium the good agreement with theory is lost. The effect was discovered during a systematic study of the SP characteristics of these films as a function of Ge overcoating thickness. The predicted linear dependence of the attenuation coefficients with Ge thickness is observed but the measured slopes, which are directly related to the plasmon frequency of the metals, are almost a factor of two larger than theory can predict. This discrepancy does not appear to be caused by absorption in the GE, interface states, surface roughness, anomalous skin effect, or transverse zero sound.

Because of the unusual new experimental technique which is being used to monitor metal-dielectric interfaces and because of the variety of new experimental measurements which have been possible, the principal investigator has been asked to give a number of invited papers on these works which are listed below:

1) "Spectroscopy with IR Surface Plamons," American Physical Society, March, 1982.

2) "IR Surface Plasmon Spectroscopy," International Conference on Ellipsometry and Other Optical Methods for Surface and Thin Film Analysis, Paris, France, June 1983.

3) "Interface Polaritons in the Infrared," International Conference on the Dynamics of Interfaces Lille, France, September, 1983.

IV. REPORTS AND PUBLICATIONS

1. "Spectroscopy with IR Surface Plasmons," A.J. Sievers, Bull. Amer. Phys. Soc. 27, 262 (1982).

2. "Dielectric Function of Silver by Surface Plasmons," H. Guggar, J.D. Swalen, A.J. Sievers, Bull. Amer. Phys. Soc. 27, 343 (1982).

3. "Influence of the Depolarization Field on Linewidth and Structure of Adsorbate Vibrational Mode Spectra," Z. Schlesinger and A.J. Sievers, Bull. Amer. Phys. Soc. 27, 410 (1982).

4. "Dipole-Dipole Coupling in Adsorbate Vibrational Mode Spectra," Z. Schlesinger and A.J. Sievers, Vibrations at Surfaces, Asilomar, California, September, 1982.

5. "IR Surface Plasmon Attenuation Coefficients for Ag and Au Films," Z. Schlesinger and A.J. Sievers, Solid State Commun. 43, 671 (1982).

6. "IR Surface Plasmon Attenuation Coefficients for Ge-coated Ag and Au Metals," Z. Schlesinger and A.J. Sievers, Phys. Rev. B26, 6444 (1982).

7. "Exploring the Optical Properties of Dielectric Coated Metals with Infrared Surface Plasmons," Z. Schlesinger, Ph.D. Thesis, 1982.

8. "Shallow Traps and the D^- center in Ge:Sb; Far-infrared photoconductivity studies below 1K," E. A. Schiff, Philosophical Mag. B 45, 69 (1982).

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